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Response surface methodology and artificial neural network analysis of crude palm kernel oil biodiesel production



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ABSTRACT

Response surface methodology (RSM) and Artificial neural network (ANN) analysis of crude palm kernel oil (CPKO) biodiesel production, using KOH and NaOH catalysts, were carried out in this research work. The four process parameters considered during the production process and modelling stages were 6–12 mol ratio of methanol/oil, 0.7–1.7 wt/wt% catalyst concentration, 48–62 °C reaction temperature and 50–90 min reaction time. Log sigmoid function and Levenberg marquardt technique were adopted in ANN while Box-Benken method was utilised for RSM. The results revealed that KOH catalyst process produced higher yield of biodiesel (87 – 99%), compared to the yield obtained from NaOH catalysed process (79 – 91%). The regression coefficients for RSM models were 0.9324 for KOH catalysed process and 0.8935 for NaOH catalysed process, while the overall correlation coefficients for ANN models were 0.82921 for KOH catalysed process and 0.89396 for NaOH catalysed process, an implication that RSM is a better analytical tool (compare to ANN) in models formulation.

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Specifications Table

| | |
|-------------------------|--|
| Subject area | Production Engineering, Chemical Engineering, Biochemical Engineering |
| Compounds | KOH, NaOH, crude palm kernel oil |
| Data category | ANN model, RSM |
| Data acquisition format | ANN model, RSM |
| Data type | Raw, analysed, simulated |
| Procedure | Experimental Design, CPKO Biodiesel Production, modelling of biodiesel yield using ANN and RSM |
| Data accessibility | Data is with the article |

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Rationale

Energy is a fundamental prerequisite for the economic advancement of every nation. This is because, every sector of a nation can only be sustained through reliable energy source. The escalating global energy demand has revealed that non-renewable energy sources are restricted wellspring of energy that will in the end run out [1–5].

In addition, the common problem associated with the utilisation of non-renewable energy sources (such as coal and crude oil) is the environmental pollution arising from the release of the associated harmful substances such as sulphur oxides (SO_x), carbon oxides (CO_x) and some hydrocarbons (HCs). Hence, there is need for alternative energy sources that are not only renewable but also sustainable in nature [6–8]. Biodiesel is a renewable form of energy source and it can be commonly produced through a process of transesterification. This process involves a reversible chemical reaction between the triglycerides of lipids (plant oils or animal fats) and the low carbon chain alcohol (methanol, ethanol, propanol) in a catalytic environment [9–15]. One of the ways to attain energy sustainability is to use non edible and/or waste cooking oils for biodiesel production. By so doing, the cost of production will reduce and the environmental challenge associated with the wrong disposal of such waste oils will be minimised [16]. Crude palm kernel oil (CPKO) is a good example of non-edible plant oil. CPKO contains 50% percent saturated fatty acids (palmitic acid $\text{C}_{16}\text{H}_{32}\text{O}_2$, lauric acid $\text{C}_{12}\text{H}_{24}\text{O}_2$ and myristic acid $\text{C}_{14}\text{H}_{28}\text{O}_2$) and 50% unsaturated fatty acid (oleic acid $\text{C}_{18}\text{H}_{34}\text{O}_2$). Crude palm kernel oil used in this study was extracted from its seeds through a crude and traditional process (at 850°C), in a local plant in Ota, Ogun state, Nigeria. [17]

According to Evangelos, the use of KOH and NaOH (as base catalysts) is commonly adopted for biodiesel production due to ease of production process, short reaction time (30 min – 2 h), low alcohol to oil molar ratio (6–12), low concentration of base catalyst (0.2 – 2.0 wt./wt.%), high yield of biodiesel (90 – 99%) and excellent catalytic performance.

Response Surface Methodology (RSM) is a collection of statistical and mathematical techniques useful for developing, improving, and optimizing processes [18 –19]. The most extensive applications of RSM are used in situations where several input variables potentially influenced some performance measure or quality characteristic of the process. Thus, performance measure or quality characteristic is called the response or yield. The input variables are sometimes called independent or process variables, and they are subject to the control or manipulation of the researcher. The field of response surface methodology consists of the experimental strategy for exploring the space of the process or independent variables, empirical statistical modelling (to develop an appropriate approximating relationship between the yield and the process variables) and the optimization methods for finding the values of the process variables that produce desirable values of the response [20]. MINITAB software is one good statistical software that can be employed for RSM.

Another tool that can be used to model a relationship between the inputs and output(s) in a statistical study is artificial neural network (ANN) [21–24]. ANN is a statistical tool in which its mode of operation of modelling formulation is based on the simulation of the structural and functionalities of biological neural networks. Basic building block of every ANN is artificial neuron, that is, a simple mathematical model (function). Such a model has three simple sets of rules: multiplication, summation and activation [25–26]. At the entrance of artificial neuron the inputs are weighted, that is, every input value is multiplied with individual weight. In the middle section of artificial neuron is sum function that sums all weighted inputs and bias. At the exit of artificial neuron the sum of previously weighted inputs and bias passes through activation function which is also called transfer function to generate response(s) [27–29]. ANN toolbox (MATLAB R2016a) is a good tool for ANN.

This research work aims at relating the analytical operations of both RSM and ANN in the CPKO biodiesel involving KOH and NaOH catalysts, by relating biodiesel yield obtained (response) to the four input variables considered. That is, the novelty of the study is in the application of RSM and ANN analytical tools to CPKO biodiesel process by establishing useful models as well as the evaluation of the level of fitness of such models.

Procedures

Materials, reagents and equipment

The materials, reagents and equipment utilised include crude palm kernel oil, NaOH, KOH, CH_3OH , H_2SO_4 (all reagents are analytical grade products of Sigma-Aldrich, Qualikems and Romil Ltd), GCMS (Agilent Technologies 7890A) and AAS (Analyst 200 Perkin Elmer precisely).

Design of experiment

Experimental design involved the use of Box-Benkhen method (Minitab 17 software) and the process parameters were methanol-oil mole ratio (6 – 12), catalyst concentration (0.7 – 1.7) wt./wt.%, reaction temperature (48 – 62) $^\circ\text{C}$ and reaction time (50 – 90) minutes, as shown in Table 1.

Biodiesel production

The biodiesel production involved transesterification reaction between CPKO and methanol, in the presence of KOH and NaOH catalysts (used separately). The procedures followed during the production were as stated in the previous work [30].

Table 1
Box-Behnken experimental design for biodiesel production.

| Parameters | Variables | Levels | | |
|-----------------------------------|-----------|--------|-----|-----|
| | | −1 | 0 | +1 |
| Methanol/Oil Mole Ratio | X_1 | 6 | 9 | 12 |
| Catalyst Concentration (wt./wt.%) | X_2 | 0.7 | 1.2 | 1.7 |
| Reaction Temperature (°C) | X_3 | 48 | 55 | 62 |
| Reaction Time (minutes) | X_4 | 50 | 70 | 90 |

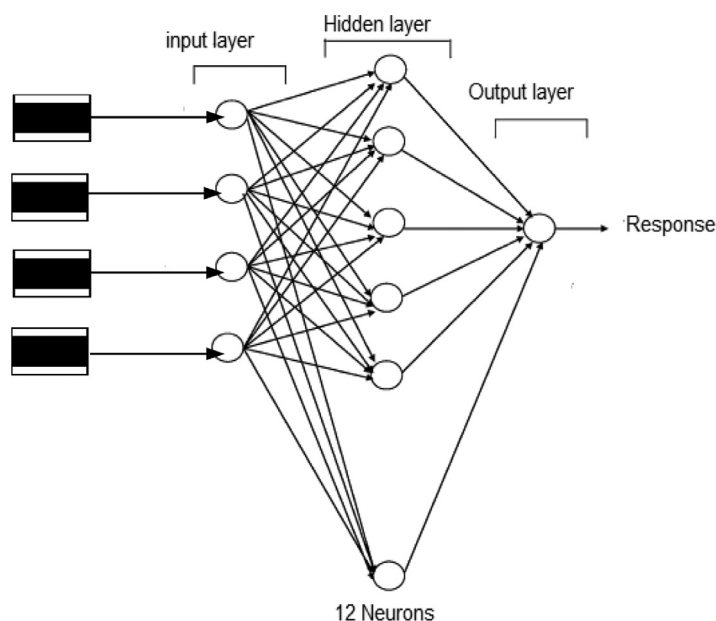


Fig. 1. ANN topology.

modelling of biodiesel yield using RSM and ANN

Using RSM, the interaction effects between the four process parameters and biodiesel yields (at the prevailing experimental conditions) were determined. modelling of biodiesel yield was carried out using RSM and ANN analytical tools, by considering four input variables (methanol/oil mole ratio, catalyst concentration, reaction temperature and reaction time) and biodiesel yield (output).

The interactive effects of the input variables on the yield was analysed and suitable models were established as well. Using ANN toolbox (MATLAB R2016a), log sigmoid function was adopted (because of its high correlation profile) for the modelling.

$$\text{logsig}(x) = 1/(1 + \exp(-x)) \quad (1)$$

The experimental data comprises of 4-input parameters such as methanol per oil mole ratio (α_m), catalyst concentration (α_c), reaction temperature (α_T) and the reaction time (α_t) as shown in Fig. 1. The input parameters were graded by dividing each column with the highest value in order to obtain values within the range of zero to one (0–1). Levenberg Marquardt technique was adopted in this work, and the training methods were sectioned into training, validation, and test set of 70%, 15% and 15% respectively. Out of the twenty-seven (27) sets of samples, 70% which corresponds to nineteen (19) samples were presented to ANN during training and the network was adjusted according to its error. Four samples (15%) were used by the network for validation purpose such that training was halted when generalization stop improving. The outstanding four samples (15%) had no effect on the training and so were used to give an independent measure of network performance during and after training.

Data, value and validation

Biodiesel yields from KOH and NaOH catalysed transesterifications

Fig. 2 shows the biodiesel yields produced from the 27 experimental runs of the transesterification of CPKO (using KOH and NaOH catalysts separately). While Fig. 3 shows the interactive effects of the four process parameters on the yields (using KOH and NaOH catalysts separately). Generally, the high yield of biodiesel could be explained in terms of the ho-

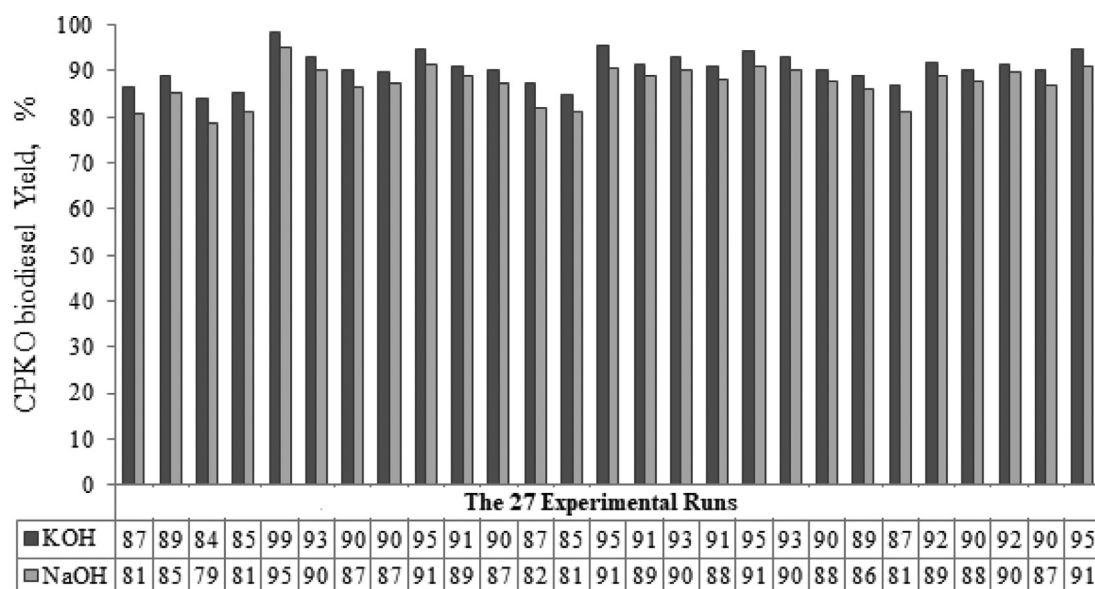


Fig. 2. CPKO biodiesel yields obtained from KOH and NaOH catalysts.

Table 2

Statistical results of the RSM model.

| Term | Coefficient | SE Coefficient | p |
|---------------------------------|-------------|----------------|-----------|
| For KOH Catalyst | | | |
| Constant | 28.5989 | 6.07910 | 0.0000000 |
| Linear | | | |
| X ₁ | 11.0532 | 0.84301 | 0.0000000 |
| X ₂ | 24.6909 | 4.06234 | 0.0000076 |
| X ₃ | −0.3862 | 0.15879 | 0.0250590 |
| Square | | | |
| X ₁ × X ₁ | −0.5788 | 0.04650 | 0.0000000 |
| X ₂ × X ₂ | −10.3358 | 1.67418 | 0.0000062 |
| X ₄ × X ₄ | −0.0034 | 0.00086 | 0.0009154 |
| Interaction | | | |
| X ₃ × X ₄ | 0.0095 | 0.00218 | 0.0003577 |
| For NaOH Catalyst | | | |
| Constant | 12.7330 | 9.04504 | 0.0000000 |
| Linear | | | |
| X ₁ | 13.3021 | 1.25430 | 0.0000000 |
| X ₂ | 26.3284 | 6.04432 | 0.000340 |
| X ₃ | −0.3182 | 0.23626 | 0.193910 |
| Square | | | |
| X ₁ × X ₁ | −0.6931 | 0.06919 | 0.0000000 |
| X ₂ × X ₂ | −11.3070 | 2.49099 | 0.000224 |
| Interaction | | | |
| X ₃ × X ₄ | 0.0083 | 0.00325 | 0.019455 |
| X ₄ × X ₄ | −0.0029 | 0.00127 | 0.033691 |

mogenous nature of all the reactants, products and catalysts. And the results generally showed that KOH catalysed transesterification process produced higher biodiesel yield compared to the yield obtained from NaOH catalysed transesterification reaction (under the same experimental conditions). Also from Fig. 3, it could be seen that the favourable conditions for high yield of biodiesel from KOH catalysed transesterification were methanol/oil mole ratio of 9.51, KOH catalyst concentration of 1.24 wt/wt%, reaction temperature of 62 and reaction time of 80 min. For NaOH catalysed reaction, favourable biodiesel yields were achieved at methanol/oil mole ratio of 9.57, catalyst concentration of 1.10 wt/wt%, reaction temperature of 62 and reaction time of 85 min. These results could be justified by the fact that KOH is chemically more active than NaOH, since lesser energy is required to excite the single valence electron in potassium than in sodium [20].

Biodiesel yield model using RSM

The results of the model formulated between biodiesel yield and the four process variables using RSM (for both KOH and NaOH catalysts) were displayed in Eqs. (2) & 3, while Table 2 revealed the statistical analysis obtained. The low values of probability value (p values below 0.05 significant level) confirmed the suitability and reliability of each of the terms

Table 3

Responses obtained from the utilisation of ANN and RSM using KOH and NaOH catalysts.

| S/N | Input parameters | | | | Yield (Response) | | | |
|-----|------------------|------------|------------|------------|------------------|-----------|----------|------------|
| | α_m | α_c | α_T | α_t | RSM KOH | ANN (KOH) | RSM NaOH | ANN (NaOH) |
| 1 | 1.00 | 1.00 | 0.89 | 0.78 | 0.87 | 0.88 | 0.81 | 0.84 |
| 2 | 1.00 | 0.41 | 0.89 | 0.78 | 0.89 | 0.88 | 0.85 | 0.87 |
| 3 | 0.50 | 1.00 | 0.89 | 0.78 | 0.84 | 0.85 | 0.79 | 0.80 |
| 4 | 0.50 | 0.41 | 0.89 | 0.78 | 0.85 | 0.86 | 0.81 | 0.82 |
| 5 | 0.75 | 0.71 | 1.00 | 1.00 | 0.99 | 0.95 | 0.95 | 0.94 |
| 6 | 0.75 | 0.71 | 1.00 | 0.56 | 0.93 | 0.94 | 0.90 | 0.87 |
| 7 | 0.75 | 0.71 | 0.77 | 1.00 | 0.90 | 0.89 | 0.87 | 0.86 |
| 8 | 0.75 | 0.71 | 0.77 | 0.56 | 0.90 | 0.93 | 0.87 | 0.88 |
| 9 | 0.75 | 0.71 | 0.89 | 0.78 | 0.95 | 0.94 | 0.91 | 0.89 |
| 10 | 1.00 | 0.71 | 0.89 | 1.00 | 0.91 | 0.91 | 0.89 | 0.88 |
| 11 | 1.00 | 0.71 | 0.89 | 0.56 | 0.90 | 0.89 | 0.87 | 0.86 |
| 12 | 0.50 | 0.71 | 0.89 | 1.00 | 0.87 | 0.87 | 0.82 | 0.82 |
| 13 | 0.50 | 0.71 | 0.89 | 0.56 | 0.85 | 0.90 | 0.81 | 0.81 |
| 14 | 0.75 | 1.00 | 1.00 | 0.78 | 0.95 | 0.97 | 0.91 | 0.87 |
| 15 | 0.75 | 1.00 | 0.77 | 0.78 | 0.91 | 0.92 | 0.89 | 0.88 |
| 16 | 0.75 | 0.41 | 1.00 | 0.78 | 0.93 | 0.94 | 0.90 | 0.91 |
| 17 | 0.75 | 0.41 | 0.77 | 0.78 | 0.91 | 0.90 | 0.88 | 0.87 |
| 18 | 0.75 | 0.71 | 0.89 | 0.78 | 0.95 | 0.94 | 0.91 | 0.89 |
| 19 | 1.00 | 0.71 | 1.00 | 0.78 | 0.93 | 0.91 | 0.90 | 0.87 |
| 20 | 1.00 | 0.71 | 0.77 | 0.78 | 0.90 | 0.89 | 0.88 | 0.88 |
| 21 | 0.50 | 0.71 | 1.00 | 0.78 | 0.89 | 0.89 | 0.86 | 0.86 |
| 22 | 0.50 | 0.71 | 0.77 | 0.78 | 0.87 | 0.87 | 0.81 | 0.79 |
| 23 | 0.75 | 1.00 | 0.89 | 1.00 | 0.92 | 0.94 | 0.89 | 0.90 |
| 24 | 0.75 | 1.00 | 0.89 | 0.56 | 0.90 | 0.91 | 0.88 | 0.87 |
| 25 | 0.75 | 0.41 | 0.89 | 1.00 | 0.92 | 0.90 | 0.90 | 0.90 |
| 26 | 0.75 | 0.41 | 0.89 | 0.56 | 0.90 | 0.93 | 0.87 | 0.88 |
| 27 | 0.75 | 0.71 | 0.89 | 0.78 | 0.95 | 0.94 | 0.91 | 0.89 |

of the two models Eqs. (2) & (3). Also, high R-sq values (close to one), as well as the low values of Sum of Errors (SE) coefficient established that the models formulated were of high accuracy, due to insignificant variation observed between the experimental data and the model-generated data. [31]

(CPKO biodiesel yield)_{KOH}

$$= 28.5989 + 11.0532X_1 + 24.6909X_2 - 0.3862X_3 - 0.5787X_1X_1 - 10.3358X_2X_2 + 0.0094X_3X_4 \\ - 0.0033X_4X_4R - Sq = 93.24\%R - Sq(adj) = 90.75\% \quad (2)$$

(CPKO biodiesel yield)_{NaOH}

$$= 12.733 + 13.3021X_1 + 26.3284X_2 - 0.3181X_3 - 0.6931X_1X_1 - 11.307X_2X_2 + 0.0082X_3X_4 \\ - 0.0029X_4X_4R - Sq = 89.35\%R - Sq(adj) = 85.43\% \quad (3)$$

Biodiesel yield model using ANN

Table 3 shows the input parameters and yields for both ANN and RSM (using KOH and NaOH catalysts). Fig. 4(a) and (b) showed the plots of ANN regression for training, validation, test and overall, using KOH and NaOH catalysts respectively, as well as their high values of regression (R close to 1). Figs. 4(a) and (b) revealed the plots of error analysis (mean squared error and error histogram) for KOH catalysed process, while Figs. 5(a) and (b) show the plots of error analysis (mean squared error and error histogram) for NaOH catalysed process.

The analysis of uni-layered architecture for mean squared error (mse) indicated an ascending order performance. The number of neurons were varied from 1 to 20 under 1000 iterations and the hidden layer suitable for this study was found at 12 neurons. While its testing of mean square error begins to diverge fast at higher neurons, the training was stopped. This research work adopted a topology of 4–12–1 because of the 4 graded input parameters, 12 hidden neurons and one response (output).

The best ANN topology occurred at the least mean squared error value of zero (0) when both the target and output have same value [31]. The method of trial and error was utilised to achieve the lowest mean square error in the validation process and the performance of the trained network as to get the response values that would replicate the target values. In this work, the best validation performance was 0.00021812 at epoch 4 for KOH catalysed CPKO process for mean squared error analysis (Fig. 5a), while the best validation performance was 0.0005605 at epoch 3 for NaOH catalysed CPKO process for mean squared error analysis (Fig. 6a). Considering the error histogram with 20 bins, the least error of -0.00231 was observed for KOH catalysed CPKO biodiesel process (Fig. 5b) while 0.000741 least error was observed for NaOH catalysed CPKO biodiesel production (Fig. 6b).

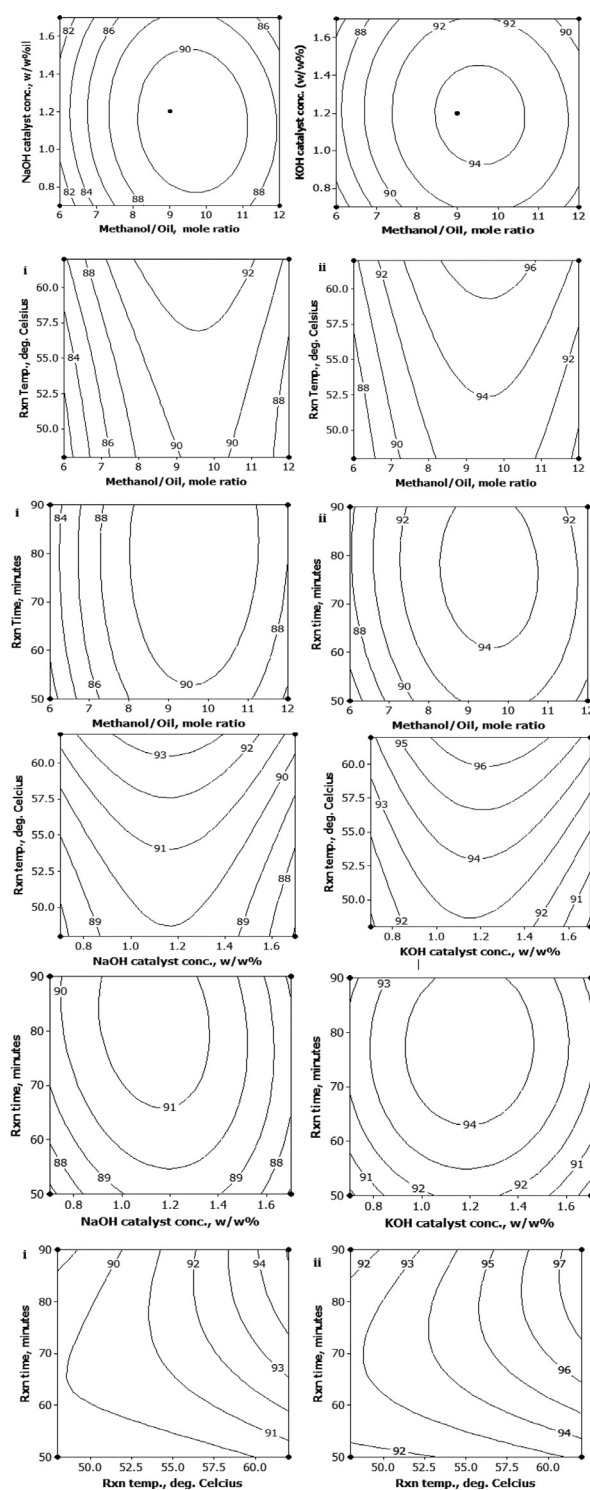
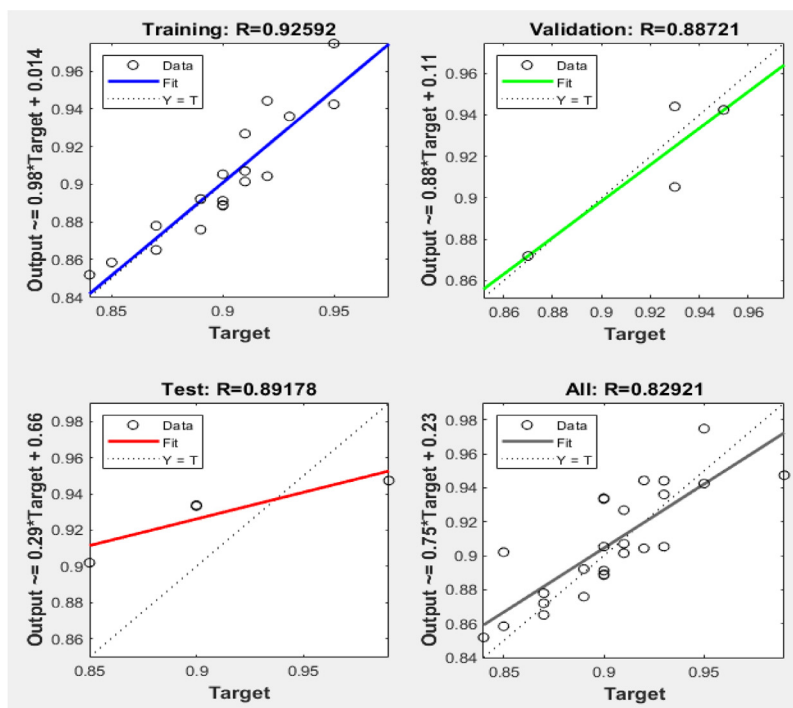
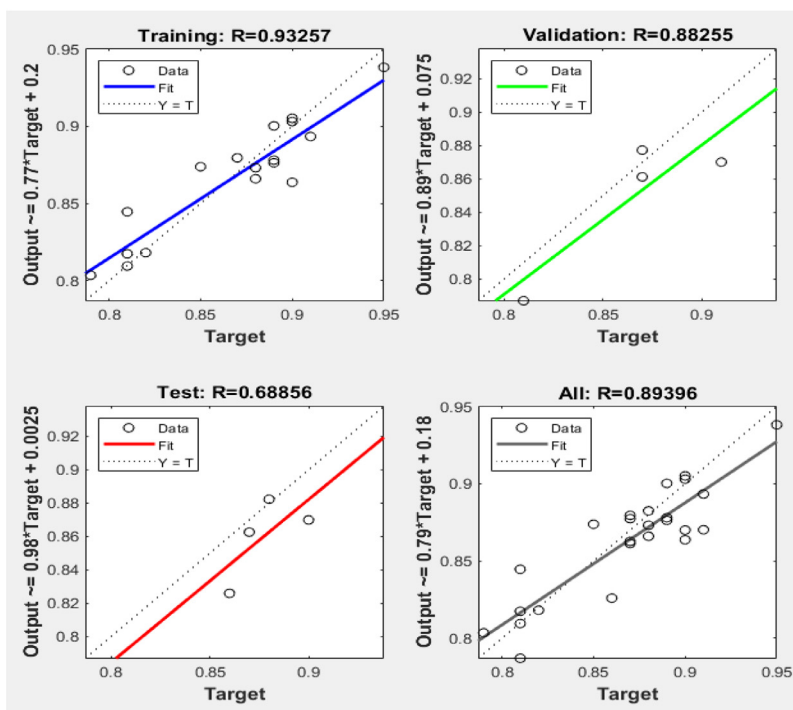


Fig. 3. Interactive effects of the process parameters on the yields (i) NaOH and (ii) KOH catalysts.

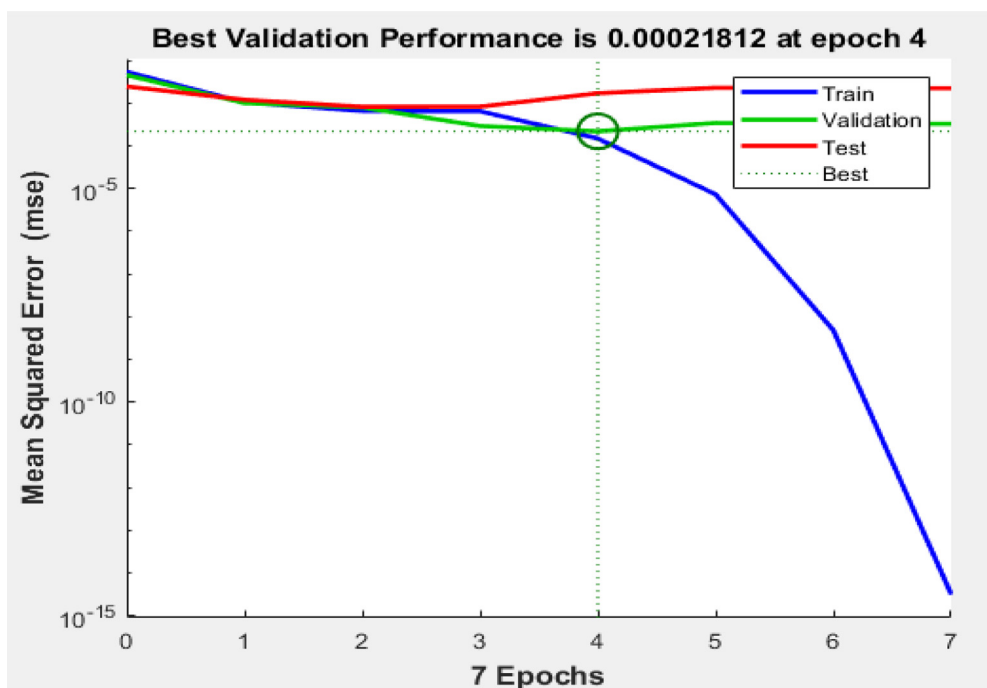


(a)

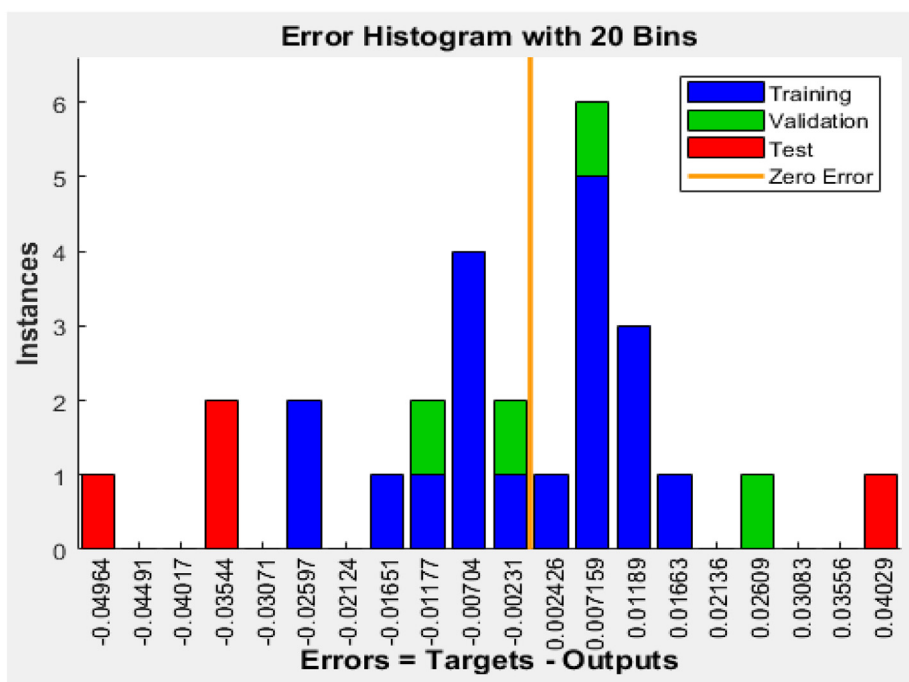


(b)

Fig. 4. Plots of ANN regression for training, validation, test and overall, using KOH catalyst Plots of ANN regression for training, validation, test and overall, using NaOH catalyst.

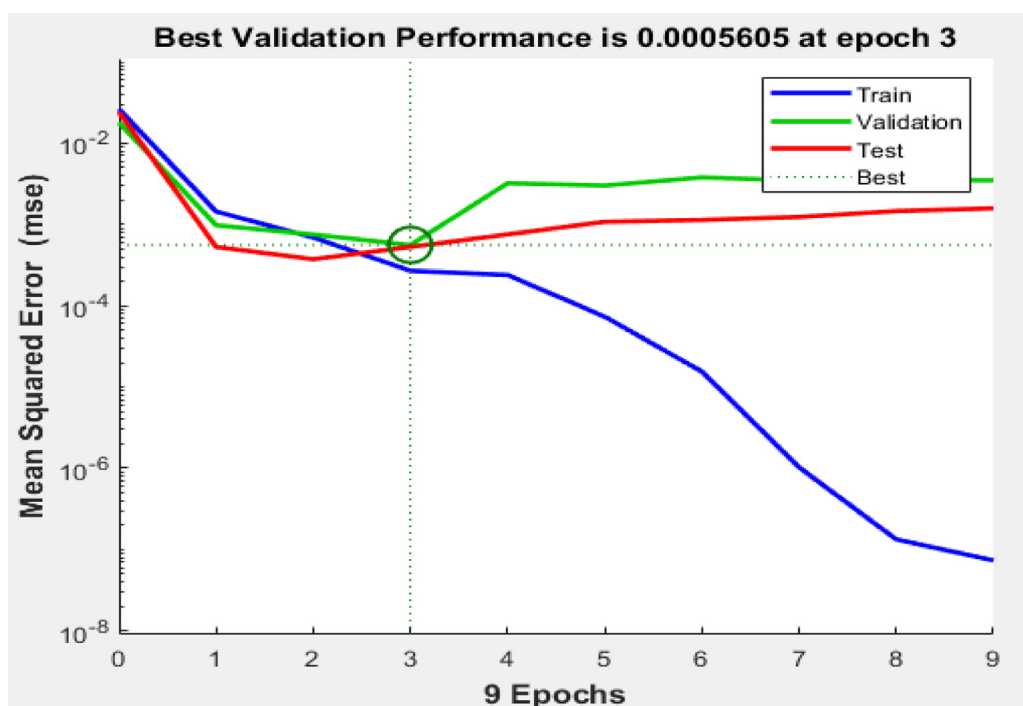


a

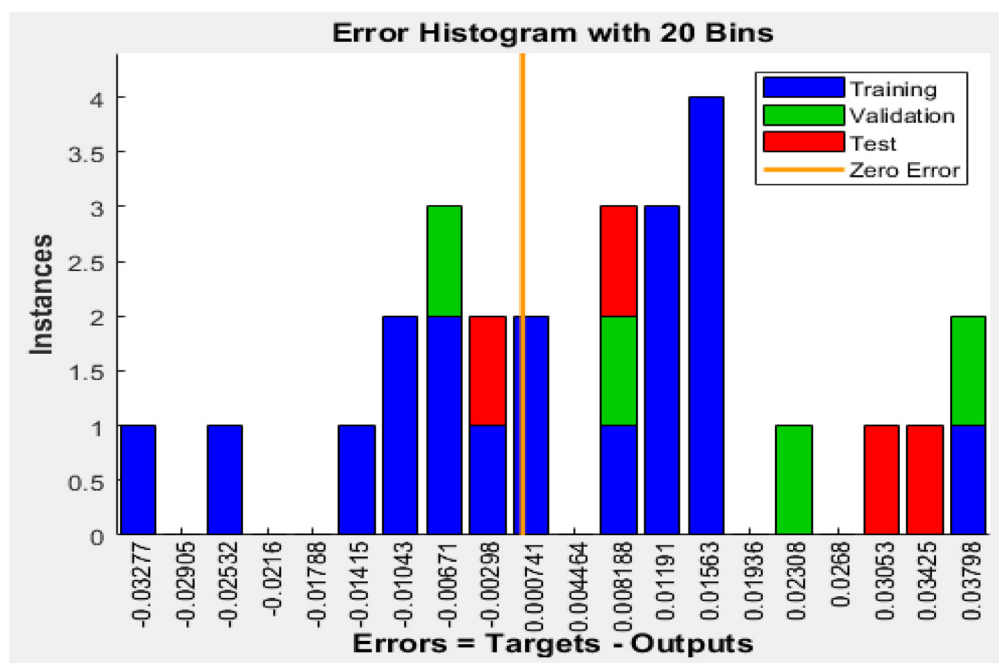


b

Fig. 5. ANN Mean squared error (mse) analysis for KOH catalysed process ANN Error histogram analysis for KOH catalysed process.



a



b

Fig. 6. ANN Mean squared error (mse) analysis for NaOH catalysed processz ANN Error histogram analysis for KOH catalysed process.

Table 4
Biodiesel properties [20].

| Property | ASTM Method | Units | CPKO Biodiesel | ASTM standard |
|------------------|-------------|--------------------|----------------|---------------|
| Density @ 25°C | ASTM D4052 | g/cm ³ | 0.8760 | 0.860 – 0.900 |
| Pour Point | ASTM D97 | °C | –6 | (–11) – (–9) |
| Flash Point | ASTM D93 | °C | 208v < 130 | |
| Water Content | ASTM D2709 | %vol. | 0.004 | ≤ 0.005 |
| Viscosity @ 40°C | ASTM D445 | mm ² /s | 4.80 – 4.90 | 1.90 – 6.00 |

Statistical comparison of rsm and ann models

The regression coefficients (R_2) for RSM models were 0.9324 for KOH catalysed biodiesel production and 0.8935 for NaOH catalysed biodiesel production process, while the overall correlation coefficients (R) for ANN models were 0.82921 for KOH catalysed transesterification process and 0.89396 for NaOH catalysed transesterification process. Also, low values of probability value (p values below 0.05 significant level) were recorded for both KOH and NaOH catalysed processes in the case of RSM (these justify the suitability and reliability of RSM models). And the low values of mean squared errors depicts the good level of accuracy of ANN models.

Properties of CPKO biodiesel produced

Table 4 shows the properties of the CPKO biodiesel obtained, as well as their ASTM standard values. The values of the properties ascertain the high quality of the biodiesel produced. This is because each value of the properties falls within the ASTM standard.

Conclusion

The research work investigated and compared the analytical strength of RSM and ANN in the transesterification process of CPKO catalysed by KOH and NaOH separately. The results of this work showed that KOH catalyst produced higher yield of biodiesel (as revealed in Fig. 2 and Table 3), due to a more reactive nature of potassium (compare to sodium). Also, the results of this research work revealed RSM as a better analytical tool (compare to ANN) in formulating models in CPKO transesterification process, as justified by the larger values of regression coefficients.

Declaration of Competing Interest

None.

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